Construction of Gas Transport Theory in Porous Media for the Entire Range of Knudsen Numbers

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論文内容要約

Chapter 1: Introduction

Gas transport through porous media has been used for various engineering devices for catalytic reaction, reductive reaction, drying and adsorption, because porous media have a large internal surface area which facilitates surface reactions. It is important to understand transport phenomena in porous media to improve the performance of such devices. Gas transport mechanism in porous media depends on the Knudsen number, which is defined as the ratio of the mean free path for intermolecular collisions of gas molecules to the pore size. In the case where the pore size of a porous medium is much larger than the mean free path for intermolecular collisions of gas molecules, i.e., in the case where the Knudsen number Kn is much smaller than unity, the gas flow through such a porous medium can be considered as a viscous continuum flow. On the other hand, in the case where the pore size of a porous medium is much smaller than the mean free path for intermolecular collisions of gas molecules, i.e., in the case where the Knudsen number is much larger than unity, the gas flow through such a porous medium is in the free molecular flow regime. In such a high Knudsen number regime, the Knudsen diffusion is the dominant transport mechanism of gas molecules through a porous medium. In the case where the pore size is comparable with the mean free path for intermolecular collisions of gas molecules, the gas flow through such a porous medium is in the transition flow regime, and hence, contributions of both Knudsen diffusion and viscous flow appear, making the problem difficult to understand. Therefore, the gas transport theory applicable to the entire range of Knudsen number has been required to clearly understand molecular transport through porous media.

Chapter 2: Construction of Theoretical Expression for Gas Transport in Micro- /Nanoscale Porous Media

In this chapter, porous media were represented by randomly arranged solid spherical particles with interpenetration of particles and pressure-driven gas flow through the porous media was simulated by using the direct simulation Monte Carlo (DSMC) method based on the Boltzmann equation. DSMC simulations were performed for different porosities and different sizes of solid particles of porous media. It was confirmed that Darcy's law holds even in the case for micro/nanoscale pores.

Furthermore, we regarded a porous medium as a bundle of tortuous circular capillary tubes, and then theoretically constructed expressions to estimate pressure-driven gas flow velocity through porous media with pores from nanoscale to microscale and porosity ranging from 0.3 to 0.5 by superposing both contributions of Knudsen diffusion and viscous flow with a velocity slip. The flow velocities estimated by using the constructed expressions agreed well with those obtained in the DSMC simulations. From the proposed expressions for flow velocity U, we also obtained the theoretical expressions that show the effect of Knudsen number on superficial velocity U_s, i.e., volume flux, permeability K and Klinkenberg coefficient b of porous media. The equations proposed here are summarized as follows:

Knudsen-type of Kawagoe-Yonemura expressions for porous media:

$$U = -\tau\left(\frac{1}{3}\lambda_{\text{wall}}\bar{C}\frac{1+c_1^{\text{K}}p}{1+c_2^{\text{K}}p}+\frac{\lambda_{\text{wall}}^2}{32\mu}p\right)\frac{1}{p}\frac{\mathrm{d}p}{\mathrm{d}x}, \qquad b = \frac{128}{3\pi}\cdot\frac{1+c_1^{\text{K}}p}{1+c_2^{\text{K}}p}\text{Kn}\cdot p,$$

$$U_{\text{s}} = -\varepsilon\tau\frac{\lambda_{\text{wall}}^2}{32}\left(1+\frac{128}{3\pi}\text{Kn}\frac{1+c_1^{\text{K}}p}{1+c_2^{\text{K}}p}\right)\frac{1}{\mu}\frac{\mathrm{d}p}{\mathrm{d}x}, \qquad c_1^{\text{K}} = 2.00\left(\frac{8}{\pi}\right)^{1/2}\left(\frac{\lambda_{\text{wall}}}{2\mu\bar{C}}\right),$$

$$K = K_0\left(1+\frac{128}{3\pi}\text{Kn}\frac{1+c_1^{\text{K}}p}{1+c_2^{\text{K}}p}\right), \qquad c_2^{\text{K}} = 2.47\left(\frac{8}{\pi}\right)^{1/2}\left(\frac{\lambda_{\text{wall}}}{2\mu\bar{C}}\right).$$

Wakao-type of Kawagoe-Yonemura expressions for porous media:

$$U = -\tau\frac{\lambda_{\text{wall}}^2}{32\mu}\left[\phi\frac{128}{3\pi}\text{Kn}+(1-\phi)\left\{1+\left(a+\frac{32}{3}\right)\text{Kn}\right\}\right]\frac{\mathrm{d}p}{\mathrm{d}x}, \qquad b = \left[\phi\frac{128}{3\pi}+(1-\phi)\left(a+\frac{32}{3}-1\right)\right]\text{Kn}\cdot p,$$

$$U_{\text{s}} = -\varepsilon\tau\frac{\lambda_{\text{wall}}^2}{32}\left[\phi\frac{128}{3\pi}\text{Kn}+(1-\phi)\left\{1+\left(a+\frac{32}{3}\right)\text{Kn}\right\}\right]\frac{1}{\mu}\frac{\mathrm{d}p}{\mathrm{d}x}, \qquad \phi = \frac{\text{Kn}}{\text{Kn}+1}.$$

$$K = K_0\left[\phi\frac{128}{3\pi}\text{Kn}+(1-\phi)\left\{1+\left(a+\frac{32}{3}\right)\text{Kn}\right\}\right],$$

where ε is the porosity of the porous medium, τ is the tortuosity of the porous medium, μ is the gas viscosity, p is the gas pressure, a is the rarefaction parameter which reflects the effect of gas rarefaction on gas viscosity, $\bar{C}$ is the mean molecular speed, λ_{wall} is the mean free path for molecule-wall collisions, i.e., the average distance traveled by a gas molecule between successive molecule-wall collisions, the Knudsen number Kn is given by $\text{Kn} = \lambda_{\text{mol}}/\lambda_{\text{wall}}$ by regarding λ_{wall} as the diameter of the tortuous tubes, λ_{mol} is the mean free path for intermolecular collisions, and K_0 represents the permeability in the continuum limit and is given by $K_0 = \varepsilon\tau\left(\lambda_{\text{wall}}^2/32\right)$.

Chapter 3: Investigation of Validity of Kawagoe-Yonemura Expressions to Different Type of Porous Media

In this chapter, we investigated whether the proposed expressions provide a good estimate of gas flow rate for any porous medium, whose internal structure is different from randomly arranged solid spherical particles. In the present study, a packed bed of spherical particles without interpenetration of particles was chosen as a different type of porous medium. It was found that when the tortuosity obtained for the randomly arranged solid spherical particles was used, our expressions underestimated the gas flow rate through the packed bed. However, when the tortuosity corresponding to the packed bed was used, the gas flow rates estimated by our expressions agreed well with those obtained in the DSMC simulations. These results suggest that a different internal structure of porous medium affects only the tortuosity in our expressions. Thus, even if a porous medium has a different internal structure from that considered in this study, our expressions may still give a good estimate of the gas flow rate by using the proper tortuosity corresponding to the porous medium.

Chapter 4: Investigation of Tortuosity of Porous Media with Nanoscale Pores Based on Paths of Moving Gas Molecules

In this chapter, the nature of tortuosity of porous medium was investigated. Tortuosity is one of the most important factors for porous medium because it determines fluid transport properties through the medium, and many tortuosity models have been proposed by empirical, analytical, and numerical approaches. As mentioned above, we constructed expressions to estimate pressure-driven gas flow rate through porous media with pores from nanoscale to microscale by regarding a porous medium as a bundle of tortuous circular capillary tubes. However, to provide a good estimate by using our expressions, the proper tortuosity corresponding to the porous medium is required. In order to evaluate the tortuosity properly, clear understanding of the meaning of the tortuosity is necessary. In the present study, we investigated the nature of tortuosity of porous medium by considering molecular paths of which gas molecules pass through it due to Knudsen diffusion. Before considering molecular paths in porous medium, we investigated molecular paths in a straight cylindrical tube in the case when intermolecular collisions are negligible. By performing numerical simulations of molecular motions in a straight cylindrical tube with diffuse reflection model for molecule-wall collisions, it was found that the ensemble average of the total path traveled by molecule until it is displaced with a certain distance is proportional to the square of the required displacement. This characteristic of the ensemble average of the total path of molecule was explained by considering the molecular motions in a straight cylindrical tube as a one-dimensional random walk. Based on this characteristic, we found that the tortuosity of porous medium can be expressed as the ratio of the average of total path traveled by molecule for a certain displacement in a straight cylindrical tube corresponding to porous medium to the average of total path traveled by molecule for the same displacement in the porous medium. Using the tortuosity estimated by this, our expressions provided a good estimate of gas flow rate for porosities less than 0.5 over the whole range of Knudsen numbers considered here.

Chapter 5: General Conclusion

In this thesis, gas transport due to the pressure gradient through porous media was investigated by numerical and theoretical method, and the theoretical expressions to estimate gas transport through porous media regardless of the Knudsen number, i.e., regardless of the pore size, were constructed. Using proposed expressions, the permeability influenced by the rarefaction effect due to the smallness of pores can be determined from the liquid permeability, which can be easily measured in experiments. These expressions will help researchers and engineers to better understand the gas transport through porous media.